IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In the matter of an application for a German Patent in the name of Merck Patent GmbH, filed under No. 101 06 198.6, and in the matter of an application for a United States Patent.

I, Dr. Ashwood Stephen DRANE, B.Sc., Ph.D., BDÜ, translator to Steve Drane Translations Ltd. of Beechwood, Chivery, Tring, Hertfordshire, England, do solemnly and sincerely declare:

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2. That I am well acquainted with the German and English languages and am a competent translator thereof.

3. That the following is to the best of my knowledge and belief a true and correct translation of the above-identified patent application and the Official Certificate attached thereto

Dated the 11th day of November 2004

Dr. Ashwood Stephen Drane

FEDERAL REPUBLIC OF GERMANY

Priority certificate regarding the filing of a patent application

File reference:	101 06 198.6
Date of filing:	10 February 2001
Applicant/proprietor:	Merck Patent GmbH, Darmstadt/DE
Title:	Plastic part comprising lustre pigments and filling bodies
IPC:	C 08 J, C 08 K, C 09 C

The attached pages are a correct and accurate reproduction of the original documents of this patent application.

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64271 D a r m s t a d t

Plastic part comprising lustre pigments and filling bodies

Print date: 11.10.2001

Plastic part comprising lustre pigments and filling bodies

The invention relates to a plastic part which may comprise lustre pigments, filling bodies and the usual assistants for plastics processing, such as pigments, stabilisers, plasticisers, fillers and reinforcing materials. Plastic parts of this type exhibit a pronounced glitter effect.

The term "lustre pigments" is taken to mean pigments in which lustre effects arise due to directional reflections at metallic or strongly light-refracting pigment particles which have a predominantly two-dimensional shape and alignment. These also include pearlescent pigments, which are lustre pigments which consist of colourless, transparent and highly light-refracting platelets. Due to parallel alignment of pearlescent pigments in plastics or in surface coatings, multiple reflections cause a soft lustre effect which is known as pearlescence. Pearlescent pigments having a certain layer thickness may satisfy the interference conditions and then exhibit iridescent colours This is where the name "interference pigments" comes from. The hue here is dependent on the viewing angle. A further feature of these transparent interference pigments is that complementary colours are visible in reflected light and transmitted light. In addition, there are also interference pigments with non-transparent support materials in which only the colour in reflected light is visible.

The lustre pigments likewise include the commercially available metal-effect pigments, such as, for example, aluminium platelets, the goniochromatic lustre pigments, based on silica platelets, aluminium platelets or iron oxide platelets as support material and the liquid-crystal pigments.

In accordance with the prior art, lustre pigments have to have a certain particle size in order that to give a glitter effect to the eye. If this is the

case, the eye is capable of recognising an individual pigment platelet as a specific particle. Since the resolving power of the eye is greatly affected by the contrast of the structures to be resolved, a specific particle size at which a glitter effect arises cannot be quoted. In general, lustre pigments having a particle size of less than 25 µm give rise to a silk to satin gloss. At particle sizes up to about 60 µm, a slightly structured sheen is observed, but it is not yet possible to refer to glittering in this case. Glitter effects are only observed at particle sizes of from 80 to 100 µm. However, particles of this size cannot always be produced, for example single crystals of basic lead carbonate or BiOCl or Al_2O_3 platelets as substrates for layer/substrate pigments can only be produced with difficulty in sizes of this order. In the case of mica pigments, the production of relatively large particles is less problematic, and pigments of this type are also commercially available.

The known lustre pigments, including the pearlescent pigments, give rise to lustre effects, but not glitter effects, in application systems, for example plastics, owing to their limited particle size.

The object of the invention is therefore to provide plastic parts in which use is made of lustre pigments which effect glitter effects although they usually only produce lustre effects owing to their particle size or which reinforce the glitter effect in pigments which already exhibit this effect.

This object is achieved by adding filling bodies which have a substantially isometric body shape to the plastic, in addition to the lustre pigment, before processing, where the diameter of these filling bodies is in the range from 10 to 200 µm. In addition, the usual assistants in plastics processing, which are described in detail in "Taschenbuch der Kunststoff-Additive; Stabilisatoren, Hilfsstoffe, Weichmacher, Füllstoffe, Verstärkungsmittel, Farbmittel" [Handbook of Plastics Additives; Stabilisers, Assistants,

Plasticisers, Fillers, Reinforcing Materials and Colorants], 2nd Edition, Carl Hauser Verlag, Munich, Vienna, may also be present.

The invention thus relates to a plastic part comprising lustre pigments comprising metallic or strongly light-refracting pigment particles or platelets which have a predominantly two-dimensional shape and alignment, and a filling body, where the filling body consists of hollow or compact particles which have a substantially isometric body shape and have a diameter of between 10 and 200 µm.

The filling bodies used are, in particular, small transparent beads, which are hollow or compact. Deviations from the ideal spherical shape are possible, but the filling bodies should predominantly have virtually isometric shapes. This is taken to mean that the dimensions of the particles are approximately the same in all three spatial directions. The surface of the filling bodies is preferably smooth. In the case of rough surfaces of the bodies, as occur, for example, in ground filling bodies, although a coarse glitter effect is observed, the colours have, for example, somewhat reduced brilliance. Preferred fillers are glass beads or hollow glass beads from the Minnesota Mining and Manufacturing Company. The refractive index of the filling body material should be approximately in the order of the refractive index of the matrix material into which the filling bodies and lustre pigments are incorporated in order to avoid excessively impairing the transparency and thus the lustre of the pigmented matrix material. However, the shape and surface area of the filling bodies are of essential importance. Of lesser importance is the material of which the filling bodies are made.

The essential parameter when selecting the filling bodies is the particle diameter of the filling bodies. This is between 10 and 200 µm, with preferred particle diameters being between 15 and 150 µm, in particular between 20 and 120 µm. The amount of filling bodies is from 0.2 to 10

parts by weight, preferably from 0.5 to 5 parts by weight and in particular from 0.5 to 3 parts by weight, based on the total weight of the plastic part.

Lustre pigments which can be employed are all appropriately known platelet-shaped pigments, such as metal-effect pigments, goniochromatic lustre pigments, interference pigments, pearlescent pigments and liquid-crystal pigments.

Metal-effect pigments consist of platelets of the metals aluminium, copper, zinc, tin and alloys thereof, in particular aluminium and gold-bronze alloys. The surface of the metal platelets has been passivated or provided with a protective layer, for example of metal oxides. Metal-effect pigments are marketed by the Eckart company under the trade name Standard®.

Goniochromatic lustre pigments consist of multicoated platelet-shaped metallic substrates, transparent non-metallic substrates or multicoated metal-oxide platelets. Aluminium platelets are employed as metallic substrate, mica is employed as transparent non-metallic substrate and iron oxide is employed as metal-oxide platelets. These lustre pigments are described in greater detail in EP 741 170, EP 708 154 and EP 753 545. They are available from BASF under the trade names Paliochrom® and Variochrom®.

Multilayered interference pigments having a light-opaque aluminium layer as the central layer are produced by the Flex company under the trade names Chromaflair® and OVP® (optically variable pigments). These pigments, which are primarily employed in security printing, basically have a five-layered structure. On a central light-opaque aluminium layer, layers of magnesium fluoride as interlayers and subsequently semi-transparent chromium layers as outer layers are deposited on both sides. The pigments are described in US 4,434,010.

Interference pigments having transparent support materials are known as pearlescent pigments. The platelet-shaped transparent support materials may be mica, other phyllosilicates, such as talc or kaolin, glass flakes, SiO_2 flakes, TiO_2 flakes or Al_2O_3 flakes. These support materials are coated with one or more metal-oxide layers. The metal oxides used here are both colourless high-refractive-index metal oxides, such as, for example, titanium dioxide or zirconium oxide, colourless low-refractive-index metal oxides, such as, for example, silicon dioxide or aluminium oxide, and coloured metal oxides, such as, for example, chromium oxide, cobalt oxide and in particular iron oxides. These platelet-shaped pearlescent pigments are known and for the most part commercially available. Pearlescent pigments are described, for example, in German Patents 31 37 808, 31 37 809, 31 51 343, 31 51 354, 31 51 355, 32 11 602 and 32 35 017. These pigments are available from Merck KGaA under the trade names Iriodin®, Colorstream® and Xirallic®.

Liquid-crystal pigments are interference pigments based on liquid-crystalline polymers. The individual pigment particles are fragments of a thin crosslinked film of liquid-crystalline polymers. The colour effects which can be achieved therewith are based on the regular structure and homogeneous arrangement of the molecules in the form of a liquid crystal and on interference, attributable thereto, of a certain spectral light fraction which is reflected by the pigment. The other light fractions pass through the pigment. Liquid-crystalline interference pigments are described in US 5,807,497 and US 5,824,733. They are available from Wacker-Chemie GmbH under the trade name HELICONE® HC.

The diameter of the particles of the non-glittering lustre pigments employed in accordance with the invention is in the range from 2 to 80 µm. The pigments can be used individually or in the form of a pigment mixture.

Pigment and filling body can likewise be added as individual components or in the form of a mixture to the matrix material, for example plastic granules. The sequence of addition of the two components is unimportant.

Lustre pigments are generally employed in plastics in concentrations of 0.5-2% by weight. In films or thin layers, significantly higher concentrations are also necessary. The crucial factor for the pigment concentration needed is always the desired effect.

The addition of filling bodies to the lustre pigments causes the latter to be deflected out of their more or less ideal alignment parallel to the surface. A certain tilt of the pigments to one another occurs, giving rise to the visual impression of individual particles having larger particle diameters than the actual particle diameters. This visual impression results in an apparent coarsening of the surface of the pigment particles tilted to one another, giving rise to a glitter effect which is observable over a much greater viewing-angle range than a glitter effect which can be achieved with lustre pigments having a large particle size.

In addition, the effects of the pigments which are larger than 80 µm are also influenced, i.e. reinforced. In the case of lustre pigments which already exhibit a glitter effect owing to their particle size, this is reinforced and likewise becomes visible over a greater angle range.

The use of the filling bodies together with strongly changing interference pigments, as available, for example, under the trade name "Colorstream®", produced by Merck KGaA, reduces the angle dependence of the colour change (flop). The transition from one colour to another colour becomes softer, i.e. the angle range within which the colour changes becomes greater.

The use of the filling bodies in the plastic part may, where appropriate, gradually reduce the visibility of flow lines and weld lines.

For the plastic part according to the invention, use can be made of known, transparent plastics, in particular thermoplastics. Suitable plastics are, for example, polyethylene (PE), polypropylene (PP), polystyrene (PS), polyphenylene oxide, polyacetal, polybutylene terephthalate, polymethyl methacrylate, polyvinyl acetate, acrylonitrile-butadiene-styrene (ABS), acrylonitrile-styrene-acrylate (ASA), polycarbonate, polyether sulfone, polyether ketones and copolymers and/or mixtures thereof. In addition, it is also possible to use casting resins, for example unsaturated polyester and methyl methacrylate casting resins. The components are incorporated into the base material in the usual manner. In the case of so-called direct colouring, the plastic granules are uniformly wetted at the surface with coupling agents, for example diisooctyl phthalate, and lustre pigments and filling bodies are then added and distributed uniformly on the surface of the plastic granules by mixing. This mixture is then processed directly in an injection-moulding machine. The mouldings obtained exhibit a very homogeneous distribution of the lustre pigments.

Direct colouring is a process which is preferably used in the laboratory. In production, masterbatches are employed. The term masterbatch is applied to a pigment preparation in which the pigment is in a significantly higher concentration than in the end product and is in fully dispersed form in a matrix which is compatible with the starting material. The incorporation of lustre pigments into plastics is described in detail in "Perlglanzpigmente" [Pearlescent Pigments], edited by Dr. Ulrich Zorll, Curt R. Vincentz Verlag, Hanover 1996.

Lustre pigments and filling bodies are incorporated into unsaturated plastics by stirring-in before the casting process.

The following examples explain the invention without representing a restriction. The formulation data regarding the lustre pigments are given in parts by weight.

Examples

Example 1

978 g of PP granules having the trade name Stamylan® PPH 10 from DSM are wetted on the surface with 2 g of diisooctyl phthalate in a tumble mixer and subsequently mixed well. 10 g of TiO_2-coated mica pigments having a particle size of from 5 to 25 µm from E. Merck, Darmstadt, Federal Republic of Germany (Iriodin 123 satin) and 10 g of filling bodies are subsequently added and distributed uniformly on the surface of the plastic granules by mixing. The filling bodies are hollow glass beads having diameters of essentially between 20 and 80 µm which are available under the trade name Scotchlite® K37 from the 3M Company, USA. The plastic granules pigmented in this way are converted into mouldings by injection moulding by means of an injection-moulding machine. Compared with Comparative Example 1, the finished mouldings exhibit significantly more coarsely structured pearlescence which does not correspond to the usual appearance in accordance with the particle size of the pearlescent pigments employed. This more coarsely structured pearlescence represents a glitter effect. The angle dependence of the lustre effect is less than in the comparative example. The proportion by weight of the pearlescent pigment and filling body in the moulding is in each case 1.

Example 2

The same components as in Example 1 are used, but the Scotchlite® K37 hollow glass beads are replaced by Scotchlite® K1 hollow glass beads from the 3M Company, USA. The particle diameter of these filling bodies is essentially in the range between 30 and 110 µm. The glitter effect observed visually corresponds to that in Example 1.

Example 3

As in Example 1, 978 g of PP granules (Stamylan® PPH10 granules) are wetted on the surface with 2 g of diisooctyl phthalate in a tumble mixer and subsequently mixed well. 10 g of a silver-coloured pearlescent pigment consisting of Al_2O_3 as substrate and a coating of TiO_2 and 10 g of filling bodies (Scotchlite® K37 hollow glass beads from the 3M Company, USA) are then added and distributed uniformly on the surface of the plastic granules by mixing. Pigments with Al_2O_3 as substrate are available under the trade name Xirallic® from Merck KGaA. The plastic granules pigmented in this way are converted into mouldings by injection moulding by means of an injection-moulding machine. The individual moulding likewise exhibits a significantly coarser, structured pearlescence than is to be expected of the pigment used owing to its particle size. This pearlescence evokes the visual impression of a glitter effect.

Example 4

983 g of PP granules (Stamylan® PPH10) are wetted on the surface with 2 g of diisooctyl phthalate and subsequently mixed well. 5 g a changing pearlescent pigment consisting of SiO_2 as substrate and Fe_2O_3 as coating and 5 g of Scotchlite® K37 hollow glass beads from the 3M Company, USA, are then added and distributed uniformly on the surface of the plastic granules by mixing. The pearlescent pigments are available under the trade name Colorstream® from Merck KGaA, Darmstadt, Federal Republic

of Germany. The proportion by weight of the pearlescent pigment and filler in the plastic part is in each case 0.5. The pigmented plastic granules are converted into mouldings by injection moulding by means of an injection-moulding machine. The injection mouldings obtained likewise exhibit a significantly coarser, structured pearlescence than would have been expected of this pigment on the basis of its particle size. The colour transition from blue-red to greenish is softer and the colour flop occurs in a broader viewing-angle range than in the case of use of the pigments without filling bodies.

Comparative Example

988 g of PP granules (available from DSM, the Netherlands, under the trade name Stamylan® PPH10) are wetted on the surface with 2 g of diisooctyl phthalate in a tumble mixer and subsequently mixed well. 10 g of lustre pigments based on TiO_2-coated mica pigments having a particle size of from 5 to 25 µm are then added (commercial product Iriodin® 123 Glanzsatin from Merck KGaA, Darmstadt, FRG) and distributed uniformly on the surface of the granules by mixing. These pigmented granules are converted into mouldings by means of an injection-moulding machine. The lustre pigment has a proportion by weight of 1 in the finished moulding. The finished mouldings exhibit good, not very structured pearlescence in accordance with their particle size.

Patent claims

1. Plastic part comprising lustre pigments comprising metallic or strongly light-refracting pigment particles or platelets which have a predominantly two-dimensional shape and alignment, and a filling body, where the filling body consists of hollow or compact particles which have a substantially isometric body shape and have a diameter of between 10 and 200 µm.

2. Plastic part according to Claim 1, characterised in that the diameters of the filling bodies are from 15 to 150 µm.

3. Plastic part according to Claim 1, characterised in that the diameters of the filling bodies are from 20 to 120 µm.

4. Plastic part according to Claim 1, characterised in that it comprises lustre pigments from the group consisting of pearlescent pigments based on mica and metal pigments coated with metal oxides, in particular titanium dioxide and/or iron oxide, and/or goniochromatic lustre pigments based on multicoated iron oxide platelets.

5. Plastic part according to Claim 4, characterised in that comprises lustre pigments based on titanium dioxide mica pigments, TiO_2- and/or Fe_2O_3-containing pearlescent pigments built up on SiO_2 platelets, TiO_2-containing, silver-coloured pearlescent pigments built up on Al_2O_3.

6. Plastic part according to Claim 1, characterised in that the proportion by weight of the filling body is from 0.2 to 10% of the total weight of the plastic part.

7. Plastic part according to Claim 6, characterised in that the proportion by weight of the filling body is from 0.5 to 5% of the total weight of the plastic part.

8. Plastic part according to Claim 1, characterised in that filling body has the shape of glass or hollow glass beads having a diameter of between 20 and 110 µm.

9. Plastic part according to Claim 1, characterised in that filling body has the shape of hollow glass beads having a diameter of between 20 and 80 µm.

Abstract

Plastic part comprising lustre pigments comprising metallic or strongly light-refracting pigment particles or platelets which have a predominantly two-dimensional shape and alignment, and a filling body, where the filling body consists of hollow or compact particles which have a substantially isometric body shape and have a diameter of between 10 and 200 µm. Mouldings of this type exhibit a pronounced glitter effect.